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The triply-ionized lanthanide rare earths (RE3+) have been widely used in the generation of coherent emission in the visible and near infrared spectral regions [1]. The active RE3+ ion is embedded in a solid state or liquid host whose ligand fields induce electric dipole transitions (ED) among the electronic levels of the $4f^n$ ground configuration. These "forced" ED transitions provide an array of optical pump bands and laser transitions throughout the lanthinide series. The primary radiation source for exploratory laser fusion implosion experiments [2] is the neodymium (Nd^{3+}) doped glass laser capable of producing energetic, subnanosecond pulses at a wavelength of 1.06 microns. Laser peak power is limited by a large nonlinear index of refraction associated with the glass host matrix; average power is limited by the low thermal conductivity of the glass host. Thus, extended use of this laser medium for the fusion application is limited primarily by the optical/mechanical properties of the glass host and not the electronic properties of the Nd^{3+} ion. This observation prompted an analysis of the radiative properties of RE3+ ions in simple molecular vapors manifesting (relatively) high vapor pressures [3]. Three specific molecular systems have been examined: rare-earth trihalides [4], rare-earth-transition-metal trihalide complexes [5], and rare-earth chealates [5]. Spectral absorption intensities have been reported in these systems for several rare-earth ions, and characterized in terms of the Judd [6]-Ofelt [7] model of "forced" ED transitions. Using this information, we have calculated the radiative transition probabilities for all 820 possible transitions in neodymium bearing vapors. The rates for transitions originating on the $^4F_{3/2}$ level and terminating on ^4I term levels are given in Table I for NdI_3, $Nd\text{-}A\ell\text{-}C\ell_3$, and ED-2 laser glass. Calculations have also been carried out for unimolecular multi-quantum vibrational relaxation rates of various excited electronic levels, which suggest high fluorescence quantum efficiency for the $^4F_{3/2}$ level in certain neodymium-bearing vapors.

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Similar calculations have been carried out for visible wavelength transitions in terbium-bearing vapors, originating on the $^5D_4$, $^5D_3$ and $^5G_6$ electronic levels. Radiative transition rates are summarized in Table II for TbI_3 vapor. Estimates of multi-quantum vibrational relaxation rates and bimolecular electronic quenching rates suggest high fluorescence quantum yields for TbI_3. Fluorsecence emission has been studied by Jacobs, et al [8] in Tb-chelate vapor and provides some insight on this question.

The exceptionally high volatility of the rare-earth -- transition-metal trihalides [5] lend themselves for storing substantial amounts of electronic energy. This is illustrated in Figure I for NdI_3 and Nd-Aℓ-$Cℓ_3$ vapors for which half of the molecules have been optically pumped.

In the presentation, the dynamical nature of these vapor systems will be discussed including rotational, vibrational, and electronic degrees of freedom. Implications of the shielded nature of the $4f^n$ electrons of the ground electronic configuration (optically active-chemically inactive) will be surveyed. Allowed 4f → 5d transitions as pump bands and ligand electronic structure will also be discussed.

TABLE I.

Radiative Transition Probabilities (A) and Transition Linestrengths (S) from $^4F_{3/2}$ J-Manifold

Terminal Level	$\lambda(\mu)$	NdI_3 S	NdI_3 A	$NdAlCl$ S	$NdAlCl$ A	Nd:ED-2 S	Nd:ED-2 A
$^4I_{9/2}$	0.88	2.6	664	1.3	340	1.2	1030
$^4I_{11/2}$	1.06	4.9	740	2.3	340	2.8	1380
$^4I_{13/2}$	1.35	2.0	140	0.8	60	1.1	270
$^4I_{15/2}$	1.88	0.3	7	0.1	3	0.2	12
ΣA_J		1550		740		2700	
τ_{rad} ($^4F_{3/2}$)		0.65		1.35		0.37	

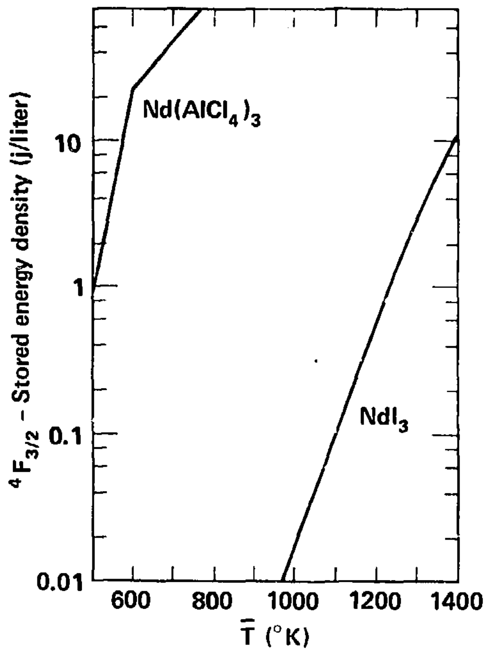
S (10^{-20} cm^2) A (sec^{-1}) τ_{rad} (msec)

TABLE II.

Radiative Transition Probabilities (A) and Transition Linestrengths (S) in TbI_3 (calc).

$^SL_J \rightarrow {^7F_{J'}}$		$^7F_0$	$^7F_1$	$^7F_2$	$^7F_3$	$^7F_4$	$^7F_5$	$^7F_6$	$\Sigma' A_J$	τ_{rad}
$^5D_4$	$\lambda(\mu)$	0.675	0.665	0.645	0.620	0.580	0.540	0.490		
	A	3	2	41	138	29	1130	79	1450	0.69
	S	1.2	0.8	14	41	7	221	12		
$^5D_3$	$\lambda(\mu)$	-	0.480	0.470	0.455	0.435	0.415	0.380		
	A	-	309	235	187	1289	216	22	2258	0.44
	S	-	33	24	17	103	15	1		
$^5G_6$	$\lambda(\mu)$	0.483	0.480	0.470	0.455	0.435	0.410	0.380		
	A	0.3	1.1	1.9	1045	1385	130	2	2565	0.39
	S	0.1	0.2	0.4	177	205	5.5	0.2		

$S(10^{-22}cm^2)$ $A(sec^{-1})$ τ_{rad}(msec)



Nd(AlCl4)3
NdI3
10
1
0.1
0.01
4F3/2 - Stored energy density (j/liter)
600
800
1000
1200
1400
T̄ (°K)

FIGURE I

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